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TRANSLATION

EFFECT OF THE MICRORELIEF OF THE CONFORMITY TO LAW OF THE DISTRIBUTION OF LIQUID METAL OVER A HARD METALLIC SURFACE

By

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The investigation of the conformity to law in the distribution of liquids over the surface of hard bodies, and in particular of the distribution of absorption-active metallic fusions over the surface of hard metals, in recent times has assumed great importance, both scientifically and practically, in connection with the study of the absorption effect of lowering the strength of solids. In considering the distribution of liquid metal over a hard metallic surface in the absence of exterior forces, including the force of gravity, it is necessary to emphasize that up to now there was basically studied only one form of such a process, namely, surface diffusion, i. e., the migration of the monolayers of molecules (atoms) of liquid from the contour of a drop applied to the hard surface and having a definite angle of contact (See, for example, [1 - 4]). Meanwhile many phenomena connected with the absorption effect, for example, the formation of long macroscopic cracks in the presence of locally applied drops of a surface-active fusion [5], accompanied qualitatively by another form of the spreading of the liquid drop—spilling. By the "spilling" we mean the elastic flow of the gradually thinning phase layer of liquid connected directly with the action of the force of gravity and with the reduction of the free surface energy of the system. In other words the condition of spreading is the condition of the full wetting of the surface.

In analysing the problem of wetting most of the authors limit themselves to a formal consideration of the relationship between the values σ_{12}, σ_{32}, and

σ_{31} (respectively the specific free surface energies of the liquid and solid on the boundary of the medium in which the experiment is performed, and on the boundary between the solid and the liquid), assuming that the full wetting is possible if

$$\sigma_{32} > \sigma_{12} + \sigma_{31}; \qquad (1)$$

if in the contrary case, i. e, with

$$\sigma_{32} < \sigma_{32} + \sigma_{31}, \qquad (2)$$

there is formed a drop with a finite angle of contact. It is necessary to emphasize, besides, that in the latter case there is possible the spreading of the liquid (by means of surface diffusion). This may go on for a considerably long time until the full disappearance of the drop applied. However, the inqualities (1) and (2) can be used for explaining the character of the spreading of the liquid only in the case of an ideally smooth surface of the solid [6 — 8]. Since in real conditions each hard surface possesses a definite microrelief characteristic of itself, for a correct description of the spreading of a liquid it is necessary, along with the physical properties of the system to take into consideration also the geometrical peculiarities of the surface. P. A. Rebinder [6] considers the additional force of friction, which acts on the contour, the amount of which is connected with the degree of roughness of the surface. This force acts as a brake on the advance of the front of the spreading drop and brings about the result that the angle of contact in the inflow is greater than with the outflow of the drop. B. V. Deryagin [7] theoretically investigated the dependence of the angle of contact on the microrelief, and came to the conclusion that with the condition $K\cos\vartheta \geqslant 1$ there can take place a spreading of a liquid over a rough surface along the micro indentations and channels (K is the coefficient of the roughness, i. e., the relationship of the true surface to the apparent one; ϑ is the angle of contact on the ideal smooth surface).

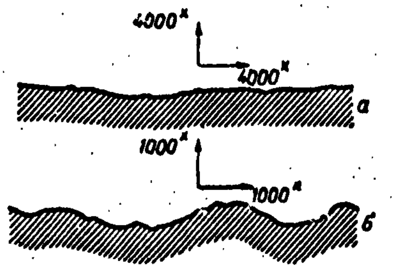

Fig. 1. Profilograms of the microrelief of zinc plates with different quality of surface: a, smooth surface (9th class of fineness): rough surface (6th class of fineness) obtained by preliminary scouring in nitric acid.

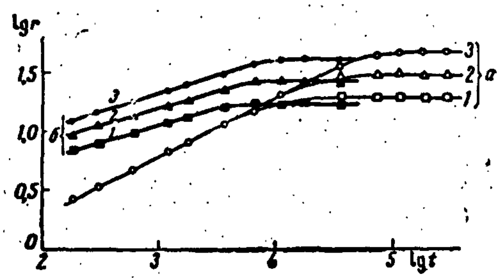

Fig. 2. Dependence of the radius of a mercury spot r (mm) on the time t (sec) with different mass of mercury m: 1, 1 mg, 2, 5 mg, 3, 10 mg: a, for smooth surface; b, for rough surface.

In the study of the spreading of mercury over a surface of semi-crystalline zinc*, free from an oxidized film, we were for the first time able to observe on one and the same liquid-solid pair** (Fig. 1) both possible forms of the spreading of a liquid, spilling over and surface diffusion. On the smooth surface (Fig. 1, a) the mercury forms a drop with an angle of contact ϑ of about 7°; from the contour of this drop there slowly spreads a round lusterless spot, the radius of which grows in accordance with the dependence $r \sim t^{0.5}$, characteristic for diffusion processes, while the mass of the drop m does not affect the shifting of the front of the spot. There is an analogous procedure in the spreading of mercury over the surface of long narrow zinc plates with one end submerged in a cup with a sufficiently great amount

*There was used for dissolving the oxidized film a 10-percent ammonium solution.

**The profilograms of the surface of the zinc plates were made in the laboratory of surface quality of the Scientific-Research Institute of the Bearing Industry.

of mercury (Fig. 3, a): it is characteristic that the speed of spreading of mercury over a smooth surface does not depend on the angle of inclination the plate to the horizontal.

The spreading of the mercury over a rough surface obtained by etching the zinc plate for 10 minutes in a 12-percent solution of nitric acid (Fig. 1, b), has qualitatively another character. The radius of the spot formed by applying a drop of mercury to the surface of the zinc in this case grows in accordance with the law $r \sim t^{0.3}$, whereby the speed of the shifting of the front of the spot is noticeably greater than in the case of surface diffusion, and it increases with the increase in the mass of mercury (Fig. 2, b). The following circumstances convincingly show that the process observed in the case in question is namely a spreading and not a two-dimensional diffusion of the mercury.

1. In applying the mercury to the rough surface there is not formed a drop with a finite angle of contace; observation of the process of spreading under the microscope confirms that the growth of the spot is accompanied by the movement of the phase layer of the mercury.

2. The speed of the rise of the mercury on the narrow strips with the rough surface increases with the lessening of the angle of inclination of the strip to the horizontal (Fig. 3, b); apparently the action of the force of gravity makes itself felt in the spreading of a phase of sufficient thickness.

3. It is possible to show that the circular spreading of the drop under the action of the force of surface tension should proceed in accordance with the law $r \sim t^{\frac{1}{4}}$ [9], which satisfactorily agrees with the experimentally observed dependence $r \sim t^{0.3}$. Actually we will accept in the first approximation that the layer of mercury in each given moment of time t has a constant thickness $z(t) = m/\pi r^2 \delta$, where m is the mass of the applied drop, $r = r(t)$

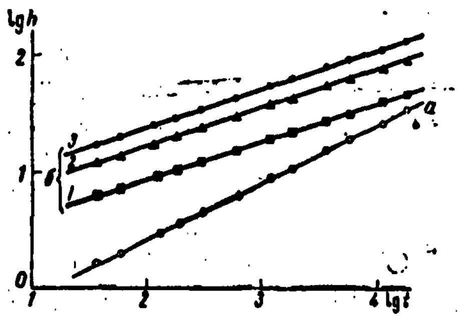

Fig. 3. Dependence of the distance h (mm) over which the mercury spread itself on the zinc plate on the time t (sec) at different angles of inclination of the specimen to the horizontal β: 1, 90°, 2, 22°, 3, 10°; a, for the smooth surface; b, for the rough surface

is the radius of the spot, δ is the density of the mercury. Then the volume of the mercury, limited by a cylinder of the radius ρ ($\rho < r$) = $V(\rho, t) = (m/\delta)\, \rho^2 / r^2$ and the average speed of the flow across the lateral surface of this cylinder amounts to $v(\rho, t) = -(1/2\pi\rho z)\partial V/\partial t = (\rho/r)dr/dt$, i. e., it increases linearly from the center of the circle to the periphery. In the case of quasi stationary viscous Neutonian flow with a gradient constant as to the thickness of the layer for the speed, the force of the viscous resistance in the elementary ring with a thickness $d\rho$ amounts to: $dF = \eta[v(\rho,t)(z/2)]\, 2\pi\rho d\rho = \eta(\rho/r)\, dr/dt\, 2\, (\pi\rho^2\delta/m)\, 2\pi\rho d\rho$ (η is the viscosity of the mercury); by integrating the latter relationship we will find the overall force which represents the flow: $F = (4\pi^2/3)\, (\eta\delta/m)\, r^4 dr/dt$. By equalling the force of the viscous resistance F to the increasing force $2\pi r\, (\sigma_{32} - \sigma_{12} - \sigma_{31}) = 2\pi r\, \Delta\sigma$, acting on the contour of the mercury film we get the equation of the motion of the front of the mercury: $(1/m)\, r^3 dr = (3/2\pi)(\Delta\sigma/\eta\delta)\, dt$. After integration we have

(3)

The relationship found not only correctly established the character of the dependence of r on t, but also enables one to explain the effect of the mass of mercury on the speed of the process (Fig. 2, b): the experimental dependence of the coefficient A on m ($A \sim m^{0.26}$) practically coincides with

the theoretical relationship $A \sim m^{\frac{1}{4}}$.

4. The difference between the spreading and the surface diffusion is very clearly brought out in the study of the effect of the temperature on the course of the distribution (Fig. 4). The rate of spreading practically does not depend on the temperature (Fig. 4, b); this is explained by the fact that the magnitudes which determine the value of the coefficient A [see equation (3)] in the temperature interval investigated change insignificantly. On the contrary the rate of the surface diffusion sharply increases with the rise in temperature (Fig. 4, a) in accordance with the temperature dependence of the coefficient of the surface diffusion: $D_n \sim \exp(-U/kT)$ (U is the energy of activation)*.

5. Assuming that on the surface of a solid there is a groove with a transverse section in the form of an isosceles triangle with an entrance angle α; then the liquid forming a drop on the surface with an angle of contact ϑ (in our case about 7°) will spread along this groove if $\vartheta < (180° - \alpha)/2$. Actually, an analysis of the profilogram of the surfaces with different degrees of roughness shows that two-dimensional diffusion gives place to spreading when the average value of the entrance angle on the surface is about equal to 160°.

In this way it is necessary to fix distinctly two qualitatively different processes of the distribution of a liquid metal over a surface of hard metal free of an oxidation film: a surface of diffusion and one of fanning out.

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*Along with the spreading of the mercury over the surface simultaneously there occurs its "soaking up" in the specimen as a result of volumetric diffusion; therefore the final value of the radius of the spot depends on the mass applied $m^{(1)0}$. With the rise in the temperature the role of the volumetric diffusion grows which leads to a decrease in the final dimensions of the spot (Fig. 4).

The surface diffusion is observed with great angles of contact; the fanning out occurs with comparatively little angles of contact and a rather considerable degree of roughness of the hard surface. Both the indicated processes observed by us in their dependence on the microrelief on the hard surface on one and the same object (mercury—zinc) are distinguished not only by the quantitative conformity to law, but they are also brought about in principle by different mechanisms.

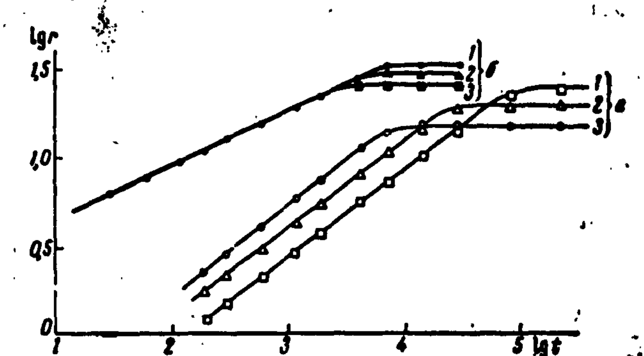

Fig. 4. Dependence of the radius of the mercury spot r (mm) on the time t (sec) at different temperatures: 1 - 0°, 2 - 20°, 3 - 40°C; a - for a smooth surface, b - for rough surface; mass of drop m = 5 mg.

In connection with what has been expounded one can express a more general proposition—it is not out of the question that the spreading or fanning out of a thin layer under the sole force of surface tension on an ideally smooth surface would not be observed at all since the suface migration of the atoms of a liquid leads to a lowering of the surface energy of a solid in the area ahead of the front of the liquid phase. In other words the full wetting in the ordinary sense of this term can prove to be unattainable in the absence of a suitable microrelief of the surface. The same phenomena can bring about also hysteresis of the wetting. In the inflow the liquid moves over the hard surface covered with monolayers of this liquid, whereas in the outflow the liquid moves already over a comparatively thin phase layer. As a result the angles of contact in the inflow and the outflow should be different. Analogous considerations can prove useful also in the analysis of the spreading of a liquid over a liquid surface.

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